

IV. "On Certain Ternary Alloys. Part VII. Alloys containing Zinc, together with Lead (or Bismuth) and Cadmium (or Antimony)." By C. R. ALDER WRIGHT, D.Sc., F.R.S., Lecturer on Chemistry and Physics in St. Mary's Hospital Medical School. Received January 5, 1893.

In the previous six papers* it has been shown that when silver is substituted for tin as "solvent" metal the critical curve deduced is uniformly *raised*, whether the two immiscible metals employed be lead and zinc, bismuth and zinc, lead and aluminium, or bismuth and aluminium; it was thought desirable to examine similarly the effects of other solvent metals, more especially *cadmium* and *antimony*, direct experiments having shown that these metals respectively are miscible in all proportions *whilst molten* with either lead, bismuth, or zinc; although in some cases more or less marked segregation of the metals from one another takes place on cooling so that solidification commences. Accordingly the experiments described below were made with zinc as lighter immiscible metal; but on substituting aluminium for zinc, so as to prepare ternary alloys containing lead (or bismuth), aluminium, and cadmium (or antimony), it was found that the close analogy between zinc and aluminium found in Part VI to subsist in alloys containing one or other of these metals along with lead (or bismuth) and tin (or silver) breaks down with these other combinations; in the case of the alloys where cadmium is the solvent metal, because molten cadmium and aluminium (contrary to the usual statements in the text-books) are *not* completely miscible together, like zinc and aluminium, but behave as lead and aluminium, or bismuth and aluminium, the heavier metal dissolving only a few tenths per cent. of aluminium, whilst this latter dissolves only some 2 or 3 per cents. of the other metal; and in the case of the alloys where antimony is the solvent, because aluminium and antimony combine together to form a most remarkable compound, represented by the formula AlSb ,† which possesses a melting point higher by upwards of 340°C . than that of the least fusible of its constituents; thus, whilst antimony melts at about 432°C ., and aluminium at below 700°C . (varying somewhat according to its purity or otherwise), the compound AlSb appears (from observations kindly made for the author by Professor Roberts-Austen with the Le Chatelier pyrometer) to have a solidifying point close to that of gold, viz., 1045° . The effect of the

* Part I, 'Roy. Soc. Proc.', vol. 45, p. 461; Part II, vol. 48, p. 25; Part III, vol. 49, p. 156; Part IV, vol. 49, p. 174; Part V, vol. 50, p. 372; Part VI, vol. 52, p. 11.

† 'Journal Society of Chemical Industry,' 1892, p. 492.

tendency towards the formation of this difficultly fusible compound with alloys richer in antimony is to cause a partial separation of antimony and aluminium thus combined together in the form of solid particles when the melted alloy is allowed to stand for some time at temperatures not exceeding some 900° ; which separation interferes with the accurate tracing out of the upper portions of the critical curve.

The discussion in detail of the results obtained with ternary alloys, containing simultaneously aluminium and cadmium (or antimony) is postponed to a future communication; but it may be here noticed that ternary alloys, such as aluminium, cadmium, and lead, or aluminium, cadmium, and bismuth, belong to a class different from that of the ternary alloys hitherto examined. Calling the three constituents A, B, and C respectively, three pairs of metals may be formed, viz., AB, AC, and BC. In the alloys previously described, *one only* of these pairs consists of two metals not miscible together in all proportions, e.g., in the case of lead, zinc, and tin, the pair lead and zinc are immiscible; whilst the pairs lead and tin, zinc and tin are perfectly miscible. With alloys of the aluminium-cadmium-lead class, on the other hand, *two* pairs of immiscible metals exist, e.g., aluminium and cadmium, and aluminium and lead. The effect of this difference is to modify very largely the nature of the critical curve deducible by means of the triangular method of graphical representation.

A priori, there seems no reason why a ternary alloy could not exist of a third class, where *all three* pairs of metal are immiscible; hitherto, however, the author has not met with such a case in actual practice. If mixed in approximately equal quantities of the three constituents, such an alloy should divide itself into *three* different ternary alloys, forming three different layers, viz., one consisting of A, with a small admixture of B and C; another chiefly containing B, with a little A and C; and a third principally consisting of C, together with a little A and B. Although metallic mixtures of this class have not been obtained so far, other sets of three liquids have been found possessing this peculiar physical character; thus a mixture of *water*, *castor oil* (genuine—not largely adulterated with other kinds of fixed oil), and *petroleum hydrocarbons* (such as ordinary kerosine) separates into three layers when well shaken up and then allowed to stand.

Alloys containing Zinc with Lead (or Bismuth) and Cadmium as Solvent.

The experiments were carried out with the lead bath arrangement in precisely the same way as before, the weighed metals being fused together with a little cyanide of potassium, and well intermixed,

poured into hot clay test-tubes, and kept 7—8 hours in the lead bath. To diminish possible volatilisation of cadmium, the temperature employed was somewhat lowered, so that it always lay between 550° and 650° , averaging near to 600° .

The analysis of the alloys containing lead, zinc, and cadmium was effected by dissolving in nitric acid and evaporating with sulphuric acid; the filtrate from the lead sulphate was diluted, further acidulated with hydrochloric acid, and treated with sulphuretted hydrogen till all cadmium was precipitated. The cadmium sulphide thus thrown down generally carried down more or less zinc sulphide; to separate this, the mixed sulphides were dissolved in a little hot concentrated hydrochloric acid, diluted, and again treated with sulphuretted hydrogen, the process being repeated when necessary, till no more zinc was contained in the filtrate. The cadmium sulphide finally obtained was dissolved in hydrochloric acid, and bromine water added in excess to destroy sulphuretted hydrogen; finally, the cadmium was precipitated boiling by sodium carbonate, and ultimately weighed as CdO . The acid liquors containing zinc were united and treated with ammonia and ammonium sulphide, the zinc sulphide being finally converted into carbonate and weighed as ZnO , correction being made for traces of Fe_2O_3 when present.

The alloys containing bismuth instead of lead were examined in the same way, excepting that the alloy was dissolved in nitro-hydrochloric acid, and the solution evaporated to dryness and treated with a large bulk of water, so as to separate the bismuth as oxychloride. This oxychloride was boiled with ammonium sulphide to remove chlorine (which is apt to interfere with proper conversion into carbonate), and the resulting bismuth sulphide dissolved in nitric acid, the solution being precipitated boiling with ammonia and ammonium carbonate, and the bismuth finally weighed as Bi_2O_3 .

Mixtures of Lead, Zinc, and Cadmium.

The following figures were obtained as the averages from the examination of 24 compound ingots (48 alloys). In the mixtures used for deducing the earlier ties, the lead and zinc were used in about equal quantities; subsequently the proportion of lead was increased relatively to the zinc, until finally the two metals were employed in about the ratio 10 to 1, this being found necessary to bring about the formation of the lighter and heavier alloys in not widely different quantities. The percentages are uniformly reckoned on the sum of the weights of the three metals found as 100.

No. of tie line.	Heavier alloy.			Lighter alloy.			Excess of cadmium percentage in lighter alloy over that in heavier.
	Cadmium.	Lead.	Zinc.	Cadmium.	Lead.	Zinc.	
0	0	98·76	1·24	0	1·14	98·86	0
1	5·17	92·85	1·98	10·12	1·54	88·34	4·95
2	13·45	83·89	2·66	24·91	2·44	72·65	11·46
3	17·57	80·18	2·25	31·48	2·85	65·67	13·91
4	19·14	78·73	2·13	36·50	3·49	60·01	17·36
5	21·44	76·34	2·22	46·42	5·96	47·62	24·98
6	22·68	75·13	2·19	55·72	10·16	34·12	33·04
7	22·23	76·32	1·45	59·20	13·44	27·36	36·97
8	22·37	76·31	1·32	70·78	18·17	11·05	48·41
9	23·70	74·85	1·45	72·76	18·80	8·44	49·06
10	33·80	64·35	1·85	72·06	21·22	6·72	38·26
11	46·93	50·82	2·25	69·50	25·86	4·64	22·57
12	50·30	47·45	2·25	67·67	28·29	4·04	17·37

Fig. 1 represents these values plotted on the triangular system, the internal dotted line being the curve obtained for the temperature 650°, with tin as solvent metal (Part V). The position of the limiting point L deduced by Stokes' second method is that where $A + A' = 73·0$, and $B + B' = 5·8$; whence—

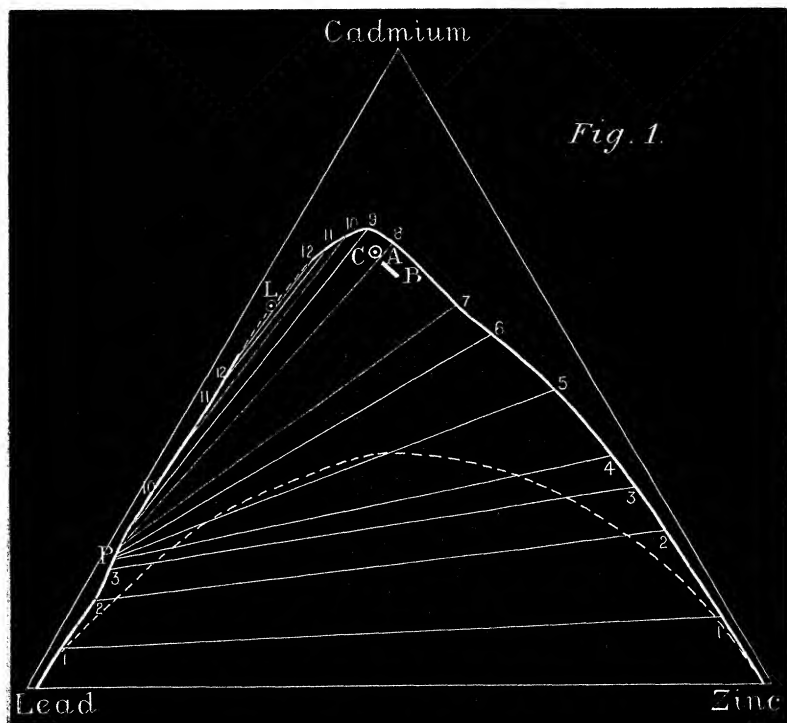
Lead	36·5
Zinc	2·9
Cadmium	60·6
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100·0	

This corresponds with a ratio between lead and zinc not far from that indicated by the formula Pb_4Zn , widely different from the corresponding ratios found when tin and silver were the solvent metals, respectively close to $PbZn_8$ and Pb_2Zn ,

The six tie lines, Nos. 4 to 9, all spring from points very close together on the left-hand branch of the curve, although the respective conjugate points on the right-hand branch are widely divergent. The centre of this cluster of points is near the point P, giving the values

Cadmium	22
Lead	76
Zinc	2
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100	

which figures represent a ratio between cadmium and lead close to that indicated by the formula Pb_2Cd .



	Calculated.	Found.	
Cd.....	21.3	22	= 22.4
Pb ₂	78.7	76	= 77.6
	100.0	98	100.0

Obviously, this configuration strongly suggests that a definite atomic compound Pb_2Cd exists, the tendency towards the formation of which is the cause of the convergence together of the tie lines; as in the analogous case with lead-zinc-tin alloys (Part V), where a similar convergence suggests the existence of the definite compound SnZn_4 .

Some observations were made at temperatures a little higher than those employed in the foregoing experiments, the result of which was to indicate that the critical curve is somewhat rapidly lowered in position with a raised temperature, at any rate, as regards its uppermost portion. Thus, at temperatures ranging between 600° and 700° , and averaging about 650° , or some 50° higher than before, it was found that no separation at all took place with a mixture containing

Cadmium	67·8
Lead	19·6
Zinc	12·6
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	100·0

This mixture is represented by the point C in Fig. 1, obviously lying distinctly *inside* the critical curve for about 600°, although necessarily *outside* that for about 650°.

Similarly, four mixtures that did separate at 650° gave points belonging to a curve lying well inside that obtained as above for about 600°; averaging the results in pairs as usual, the following values were obtained :—

Heavier alloy.			Lighter alloy.		
Cadmium.	Lead.	Zinc.	Cadmium.	Lead.	Zinc.
22·11	76·44	1·45	63·96	19·59	16·45
22·72	74·98	2·30	67·35	18·85	13·80

The line AB represents the portion of the right-hand branch of the critical curve thus traced out at about 650°, obviously lying well inside that found at near 600°; the corresponding points on the left-hand branch lie close to the point P, approximately corresponding with the ratio Pb₂Cd.

Mixtures of Bismuth, Zinc, and Cadmium.

A series of twenty compound ingots (forty alloys) gave the following average results; in the earlier cases the bismuth and zinc were employed in equal quantities; in the later ones the proportion of zinc was increased up to 1½ times the bismuth :—

No. of tie line.	Heavier alloy.			Lighter alloy.			Excess of cadmium percentage in lighter alloy over that in heavier.
	Cadmium.	Bismuth.	Zinc.	Cadmium.	Bismuth.	Zinc.	
0	0	85.72	14.28	0	2.32	97.68	0
1	6.98	78.84	14.18	3.77	3.32	92.91	- 3.21
2	14.27	71.34	14.39	9.80	4.46	85.74	- 4.47
3	21.19	64.38	14.43	14.37	5.54	80.09	- 6.82
4	28.24	56.75	15.01	18.04	6.02	75.94	-10.20
5	33.67	50.34	15.99	24.48	6.34	69.18	- 9.19
6	38.06	46.02	15.92	29.32	6.90	63.78	- 8.74
7	46.20	37.08	16.72	37.42	7.01	55.57	- 8.78
8	53.88	24.93	21.19	46.73	9.07	44.20	- 7.15
9	54.48	21.21	24.31	49.73	12.29	37.98	- 4.75
10	53.08	21.55	25.37	50.93	15.25	33.82	- 2.15

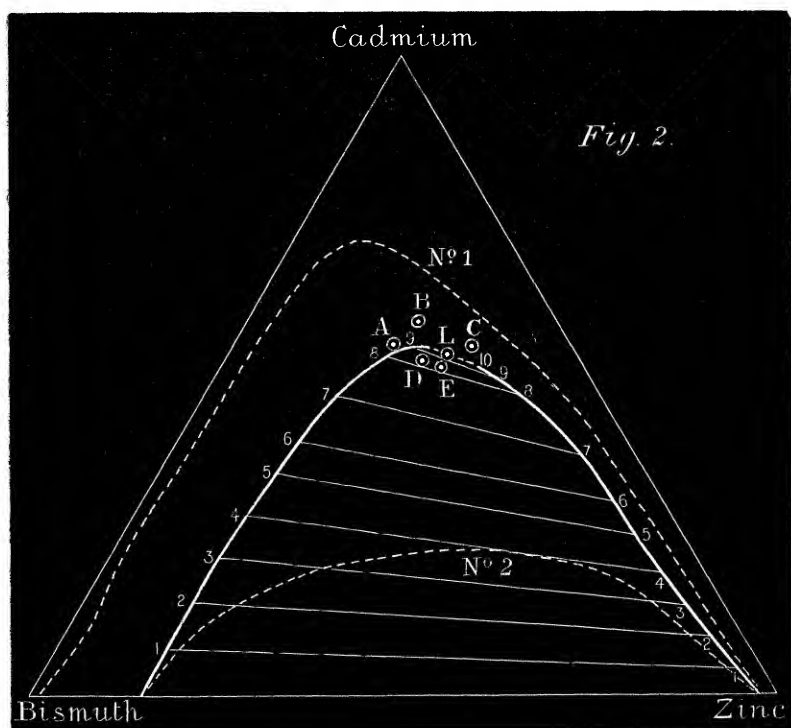


Fig. 2 represents these values plotted on the triangular system; the outer dotted line (No. 1) being the curve above described obtained

with lead, zinc, and cadmium, and the inner one (No. 2) that obtained at 650° with bismuth, zinc, and tin (Part V). The position of the limiting point L is deduced by Stokes's 2nd method, as that where $A + A' = 39$ and $B + B' = 57$, whence

Bismuth.....	19.5
Zinc	28.5
Cadmium	52.0
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	100.0

This corresponds with a ratio between bismuth and zinc close to that indicated by the formula BiZn_5 ; the corresponding ratios previously found with tin and silver as solvent metals being respectively close to BiZn_{10} and BiZn_2 .

The three points marked A, B, C, lying *outside* the critical curve, represent the compositions of three mixtures that did not separate at temperatures lying near to 600° , the mean temperature corresponding with the critical curve delineated; on the other hand, the points D and E, lying *inside* the critical curve, represent two mixtures that did not separate when the temperature was somewhat raised (to near 650°); showing that with mixtures of bismuth, zinc, and cadmium, as with those of lead, zinc, and cadmium, a considerable depression of the upper part of the critical curve is brought about by a comparatively slight elevation of temperature.

	A.	B.	C.	D.	E.
Bismuth	23.34	20.29	18.71	21.50	21.05
Cadmium	54.49	57.38	53.89	52.20	51.00
Zinc	22.17	22.33	27.40	26.30	27.95
	<hr/>	<hr/>	<hr/>	<hr/>	<hr/>
	100.00	100.00	100.00	100.00	100.00

On contrasting together the two critical curves thus obtained with cadmium as solvent it is noticeable that in this case also the same rule is observed as was found to hold in all the previously-described cases, viz., that, *ceteris paribus*, the substitution of bismuth for zinc as the heavier of the two immiscible metals depresses the curve. As regards the direction of slope of the tie lines, however, it is remarkable that with cadmium as solvent the ties slope uniformly to the *left* with lead, and to the *right* with bismuth, as the heavier immiscible metal. With silver as solvent all ties slope to the *left* whether lead or bismuth be the heavier immiscible metal, and whether zinc or aluminium be the lighter one; whilst with tin as solvent the opposite

is the case, the ties here all sloping to the *right*, with the exception of the lower ties in the case of the mixtures lead-tin-zinc and lead-tin-aluminium; this exception being presumably due, as previously described, to the influence of the tendency towards the formation of a definite compound of lead and tin, Pb_3Sn .

Alloys containing Zinc with Lead (or Bismuth) and Antimony as Solvent.

These experiments were made in the same way as before, the only difference being that the temperatures at which the mixtures were kept in tranquil fusion lay between 600° and 700° , averaging near to 650° .

The resulting alloys were analysed by dissolving in diluted aqua regia, precipitating by sulphuretted hydrogen (after copious further dilution), separating antimony from lead (or bismuth) sulphide by repeated treatment with ammonium sulphide, and finally collecting on a weighed filter the mixed sulphur and antimony sulphide thrown down on acidulating the filtrate, and heating a known fraction of the dried precipitate in a current of carbon dioxide so as to expel sulphur and leave antimony sulphide. In the case of lead alloys the lead sulphide left undissolved was dissolved in nitric acid, evaporated with sulphuric acid, and the lead sulphate finally obtained weighed as such; when the acid filtrate from this contained small quantities of zinc (as occasionally happened, owing to zinc sulphide being carried down along with lead and antimony sulphides) this liquid was added to the first zinc-containing filtrate, and the two jointly precipitated with ammonia and ammonium sulphide, the zinc being ultimately transformed into carbonate, and weighed as ZnO as usual, correction being made for traces of Fe_2O_3 when present. In the case of bismuth alloys, the bismuth sulphide left undissolved by ammonium sulphide was dissolved in nitric acid, and precipitated boiling by ammonium carbonate. When zinc was present (carried down as sulphide, as before) this was chiefly found in the ammonium carbonate filtrate; sometimes, however, traces were carried down with this basic bismuth carbonate; these were separated by dissolving the weighed impure Bi_2O_3 in hydrochloric acid, evaporating to dryness, diluting the residue largely with water, and filtering off from the precipitated bismuth oxychloride.

In all cases the analyses were calculated taking the sums of the weights of the three metals found as 100.

Mixtures of Lead, Zinc, and Antimony.

The following average values were deduced from fourteen compound ingots (twenty-eight alloys), the proportion between lead and zinc throughout being near to equality:—

No. of tie-line	Heavier alloy.			Lighter alloy.			Excess of antimony percentage in lighter alloy over that in heavier.
	Antimony.	Lead.	Zinc.	Antimony.	Lead.	Zinc.	
0	0	98.76	1.24	0	1.14	98.86	0
1	2.69	93.87	3.44	4.60	2.27	93.13	+ 1.91
2	7.11	87.02	5.87	11.41	7.53	81.06	+ 4.30
3	8.37	85.22	6.41	16.75	13.52	69.73	+ 8.38
4	9.77	80.81	9.42	22.33	17.46	60.21	+ 12.56
5	15.45	67.17	17.38	26.68	24.34	48.98	+ 11.23
6	21.00	53.65	25.35	27.11	29.55	43.34	+ 6.11
7	22.80	48.48	28.72	28.15	30.36	41.49	+ 5.35

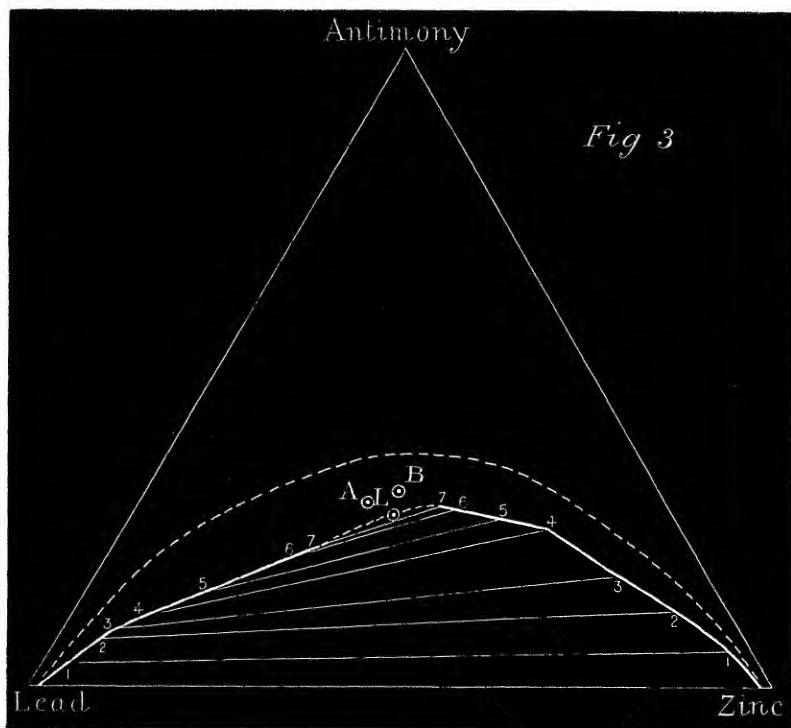


Fig. 3 represents these values plotted on the triangular system, the exterior dotted line indicating the corresponding curve obtained with tin as solvent instead of antimony (Part V). The points marked A and B represent two alloys that did not separate, containing

	A.	B.
Antimony	28·5	30·8
Lead	41·5	32·2
Zinc	30·0	37·0
	<hr/> 100·0	<hr/> 100·0

By means of Stokes' first method the values for the limiting point are deduced as being $A-B = 4$, $C+C' = 53$; whilst by means of the 2nd method the following values are found, $A+A' = 75$, $B+B' = 72$; leading to the final result

	1st Method.	2nd Method.	Mean.
Lead.....	38·75	37·5	38·1
Zinc.....	34·75	36·0	35·4
Antimony ..	26·50	26·5	26·5
	<hr/> 100·00	<hr/> 100·0	<hr/> 100·0

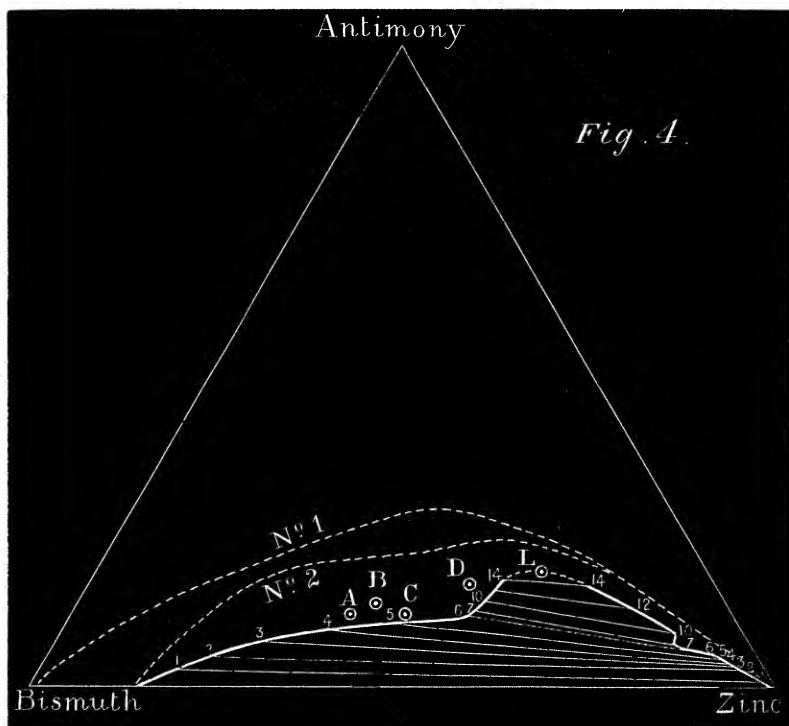
This indicates a ratio between lead and zinc near to that represented by the formula $PbZn_3$; the corresponding ratios found with tin, silver, and cadmium as solvent metals being respectively near to $PbZn_6$, Pb_2Zn , and Pb_4Zn .

Mixtures of Bismuth, Zinc, and Antimony.

The following average values were deduced from the examination of twenty-eight compound ingots (fifty-six alloys); in the earliest cases the ratio of zinc to bismuth was that of equality; later it was raised successively to 2 : 1 and 3 : 1, and finally to 7 : 2:—

No. of tie line.	Heavier alloy.			Lighter alloy.			Excess of antimony percentage in lighter alloy over that in heavier.
	Antimony.	Bismuth.	Zinc.	Antimony.	Bismuth.	Zinc.	
0	0	85·72	14·28	0	2·32	97·68	0
1	1·55	81·23	17·22	0·43	3·23	96·34	-1·12
2	4·10	70·02	25·88	2·15	4·75	93·10	-1·95
3	6·33	60·67	33·00	3·09	5·07	91·84	-3·24
4	8·63	53·00	38·37	3·67	6·02	90·31	-4·96
5	10·03	44·05	45·92	4·32	8·02	87·66	-5·71
6	10·90	37·09	52·01	5·63	10·11	84·26	-5·27
7	11·74	35·05	53·21	5·54	11·26	83·20	-6·20
8	12·84	33·03	54·13	5·69	10·47	83·84	-7·15
9	13·22	33·20	53·58	6·47	9·76	83·77	-6·75
10	14·12	32·66	53·22	7·02	8·65	84·33	-7·10
11	15·87	30·67	53·46	9·25	10·37	80·38	-6·62
12	16·04	28·61	55·35	12·81	15·19	72·00	-3·23
13	16·75	27·66	55·59	14·43	17·56	68·01	-2·32
14	17·00	26·58	56·42	15·53	18·28	66·19	-1·47

Fig. 4 represents these values, the outer dotted line, No. 1, indicating the corresponding curve for lead-zinc-antimony alloys, and that marked No. 2 the similar curve for bismuth-zinc-tin alloys (Part V), all referring to the same average temperature of 650° , or thereabouts.



The points marked A, B, C, D indicate four mixtures that did not separate; viz. :—

	A.	B.	C.	D.
Antimony.....	11·71	14·6	11·6	15·63
Bismuth.....	50·17	42·7	44·2	30·89
Zinc.....	38·12	42·7	44·2	53·48
	100·00	100·0	100·0	100·00

By means of Stokes' second method, the limiting values are deduced, $A + A' = 45·5$, $B + B' = 119·5$; whence the composition at the limiting point is—

Antimony.....	17.50
Bismuth.....	22.75
Zinc.....	59.75
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	100.00

Here the bismuth and zinc are in proportions near to those indicated by the formula BiZn_8 ; with tin, silver, and cadmium as solvent metals the corresponding proportions were respectively near to those indicated by BiZn_{10} , BiZn_2 , and BiZn_5 .

It is noticeable that the limiting point thus deduced is the highest point of the entire curve; whereas, in every one of the other eleven curves so far investigated, the limiting point lies on one side or the other of the highest point, and more or less lower down.

Two remarkable irregularities are visible in the contour of the above-deduced critical curve for antimony-bismuth-zinc alloys; on the right-hand side a peculiar notch or depression is noticeable, strongly suggesting a phenomenon similar in character to that previously observed in the case of silver-zinc-lead and silver-zinc-bismuth alloys; *i.e.*, the formation of a definite compound of solvent with one of the two immiscible metals more soluble in the other of these two metals than any other neighbouring mixture of the two in other proportions. On the left-hand side an analogous, but far wider, depression is observable. These depressions reach their greatest depths at the 7th tie line, where at each of the two conjugate points the ratio between the bismuth and antimony present is not far from that corresponding with the formula Bi_3Sb_2 .

	Left hand (heavier alloy).		Calculated for Sb_2Bi_3 .		Right hand (lighter alloy).	
Antimony..	11.74	= 25.1	27.8		5.54	= 32.9
Bismuth...	35.05	= 74.9	72.2		11.26	= 67.1
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	46.79	100.0	100.0		16.80	1.000

Experiments now in progress indicate that an analogous depression exists in the critical curve obtained with antimony-bismuth-aluminium alloys, also most strongly marked at a point where the bismuth and antimony are in proportions not far from that corresponding with Bi_3Sb_2 .

On comparing together the two curves deduced with antimony as solvent it is noticeable that they show the same general characters as the corresponding pair of curves similarly obtained with cadmium as solvent; *i.e.*, whilst the curve with bismuth as heavier immiscible metal lies *inside* that with lead (as in all other similar cases as yet examined), the direction of slope of tie lines is opposite in the two

cases; viz., uniformly to the *right* with *bismuth* and to the *left* with *lead*. It is remarkable, however, that this relationship does not hold when aluminium is substituted for zinc, the experiments now in progress indicating that the ties then *always slope to the left*, whether lead or bismuth be the heavier immiscible metal.

On comparing together the three sets of critical curves deduced for temperatures not far apart (600—650°) with tin, cadmium, and antimony respectively as solvent metal, it is noticeable that whether bismuth or lead be the heavier immiscible metal the curve with cadmium as solvent lies *outside*, and that with antimony *inside*, the curve deduced with tin as solvent. The curves obtained with silver as solvent cannot properly be directly compared with these on account of the higher temperatures (800—870°) employed; but, judging from the marked effect of a rise of temperature in depressing the critical curves obtained with cadmium as solvent, it seems probable that for the same temperature the curve with silver as solvent would be found to lie *outside of that with cadmium as solvent*, the two immiscible metals being the same. At any rate, in all cases the curve with silver as solvent lies far outside that similarly obtained with tin as solvent.

In the case of alloys containing aluminium as lighter immiscible metal, it has been shown (Part VI) that with silver as solvent the critical curve also lies *outside* that obtained with *tin* as solvent, whether bismuth or lead be the heavier immiscible metal. The experiments now in progress seem to indicate that the corresponding curves with antimony as solvent lie again *inside* the curves deduced with tin as solvent. As already stated, corresponding curves with cadmium as solvent cannot be obtained, as the immiscibility of aluminium and cadmium causes the resulting ternary alloys to belong to an entirely different class, the critical curves pertaining to which cannot be directly compared with those belonging to ternary mixtures analogous to the twelve so far investigated.

Much of the analytical work requisite for the above experiments was carried out by Mr. Sydney Joyce, to whom the author's acknowledgments are due for the assistance rendered.

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Transactions.

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The Society.

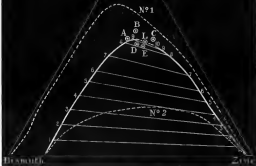
Cardium

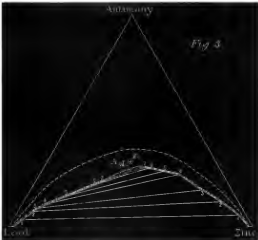
Fig. 1



Euclytium

Fig. 2





Antimony

Fig. 4.

Bismuth

Zinc